

Susceptibility inhomogeneity and non-Fermi liquid behavior in nominally ordered UCu_4Pd

D. E. MacLaughlin

*MS K764, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
Department of Physics, University of California, Riverside, California 92521-0413*

R. H. Heffner

MS K764, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

G. J. Nieuwenhuys

Kamerlingh Onnes Laboratory, Leiden University, 2300 RA Leiden, the Netherlands

G. M. Luke, Y. Fudamoto, Y. J. Uemura

Department of Physics, Columbia University, New York, NY 10027

R. Chau,* M. B. Maple

Department of Physics, University of California, San Diego, La Jolla, California 92093

B. Andraka

Department of Physics, University of Florida, Gainesville, Florida 32611

(February 1, 2008)

Muon spin rotation experiments on a stoichiometric sample of the non-Fermi liquid (NFL) heavy-fermion compound UCu_4Pd , in which recent neutron scattering experiments suggest an ordered structure, indicate that the U-ion susceptibility is strongly inhomogeneous at low temperatures. We argue that this is due to residual disorder, which also dominates NFL behavior. The data yield a short correlation length ($\lesssim 1$ lattice spacing) and a rapid low-temperature U-moment relaxation rate ($\gtrsim 10^{12} \text{ s}^{-1}$), which constrain cluster-based models of NFL behavior.

PACS numbers: 71.27.+a, 75.30.Mb, 76.60.Cq.

Many heavy-fermion alloys and compounds containing f atoms have been found to exhibit thermodynamic and transport properties at low temperatures which are not in agreement with the conventional Landau Fermi-liquid predictions.¹ Most such non-Fermi liquid (NFL) materials share two characteristics: proximity to a magnetic region of the appropriate phase diagram, and disorder due to chemical substitution. A number of mechanisms, some of which include one or both of these features, have been taken to be the basis for NFL behavior. This paper presents strong evidence for a disorder-driven NFL mechanism in nominally ordered UCu_4Pd .

Nuclear magnetic resonance² (NMR) and muon spin rotation^{3,4} (μSR) experiments in $\text{UCu}_{5-x}\text{Pd}_x$ alloys, which exhibit NFL behavior for $1 \lesssim x \lesssim 1.5$ (Ref. 5), revealed static linewidths which vary more rapidly than the bulk susceptibility with temperature and which become anomalously large at low temperatures. This is unambiguous evidence for strong inhomogeneity of the local magnetic susceptibility, which in turn suggests a large effect of disorder on the electronic structure of these alloys. This article describes μSR measurements in a new sample of UCu_4Pd , for which recent neutron diffraction experiments⁶ have been interpreted as con-

sistent with crystalline order. We find excellent agreement with the original μSR results, again indicating considerable susceptibility inhomogeneity. Either there is a residual level of disorder in this sample, which is possible given the uncertainty in the neutron diffraction results, or UCu_4Pd exhibits a novel periodic spatial modulation of the local U-moment susceptibility in the absence of long-range spin order. We will argue that the former is most likely. We also find that the susceptibility inhomogeneity is characterized by a short correlation length (of the order of a lattice constant), and that the U moments fluctuate at a rate $\gtrsim 10^{12} \text{ s}^{-1}$. These latter results pose constraints for cluster-based theories of disorder-driven NFL behavior.

Hybridization between conduction-electrons and local moments gives rise to Kondo and other correlations in heavy-fermion systems. Several treatments have appeared which ascribe NFL behavior to disorder in this hybridization. Bernal *et al.*² proposed a simple phenomenological “Kondo disorder” model, in which the disorder results in a wide distribution of Kondo temperatures T_K . At a given temperature T local moments with $T_K < T$ are uncompensated and give rise to the NFL behavior. The wide spread of Kondo temperatures gives rise to a

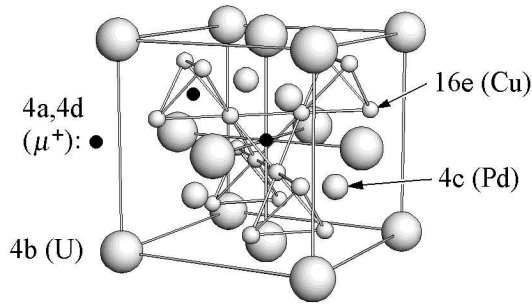


FIG. 1. Unit cell of UCu₄Pd assuming structural order, with atom sites indicated in Wyckoff notation. Muon stopping sites in the end compound UCu₅ (Ref. 11) are also shown.

temperature-dependent spread $\delta\chi$ of local-moment susceptibilities $\chi(T, T_K)$. A similar picture was proposed independently by Matsuhira *et al.*⁷ in connection with their experiments on randomly diluted CeRu₂Si₂. Miranda *et al.*⁸ put this model on a firmer theoretical foundation, and pointed out that it was capable of explaining the ubiquitous linear temperature dependence of the low-temperature electrical resistivity.

Recently Castro Neto *et al.*⁹ have extended the simple single-impurity Kondo disorder model to take into account the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling between local moments. They concluded that the combination of disorder and a critical point associated with cooperative behavior of these local moments give rise to a Griffiths phase of correlated clusters associated with a nonuniversal scaling exponent λ . This theory is in good agreement with specific heat and susceptibility data for a number of NFL materials.¹⁰

Disorder is an essential ingredient for these NFL mechanisms. Some NFL compounds may crystallize in ordered lattice structures, however, in which case there is no reason to suspect a disorder-driven mechanism for NFL behavior if the atomic order is perfect. UCu₄Pd is a candidate for such an ordered compound.

UCu_{5-x}Pd_x, $0 \leq x \lesssim 2.5$, crystallizes in the fcc AuBe₅ structure (space group $F\bar{4}3m$). The end compound UCu₅ possesses two crystallographically inequivalent copper sites in the ratio 4 : 1 at the 16e and 4c positions (Wyckoff notation). Thus stoichiometric UCu₄Pd could order as shown in Fig. 1. The concentration dependence of the lattice parameter exhibits a break at $x \approx 1$ (Ref. 5), which suggests preferential Pd occupation of 4c sites but does not determine the degree of order.

Recently Chau *et al.*⁶ reported elastic neutron diffraction measurements on members of the UCu_{5-x}Pd_x series. From Rietveld refinements of the data they find that indeed Pd atoms preferentially occupy 4c sites for $x < 1$,

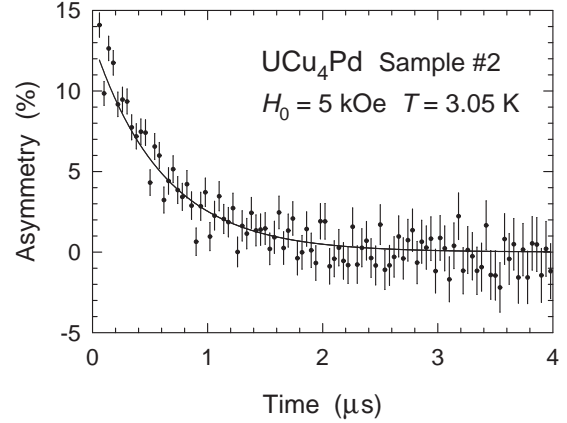


FIG. 2. Representative TF- μ SR asymmetry relaxation function (after transformation to a reference frame rotating with the μ^+ precession frequency) in UCu₄Pd sample (Sample #2) used in neutron diffraction studies (Ref. 6). Curve: best-fit exponential.

and a mixture of Cu and Pd atoms occupy 16e sites for $x > 1$. For $x = 1$ “...there is no evidence for Pd/Cu disorder...”.⁶ The data do not rule out the possibility of site interchange between Pd and Cu sites, at the level of $\sim 4.3\%$ occupation of 16e (Cu) sites by Pd atoms [and therefore $\sim 17\%$ occupation of 4c (Pd) sites by Cu atoms]. A minimum in the variation with x of the residual resistance ratio (RRR) = $R(300\text{ K})/R(1\text{ K})$ was also observed near $x = 1$, indicating that the electronic structure undergoes a transition near this Pd concentration.

In view of these results we have carried out μ SR studies on a portion of the same powder sample of UCu₄Pd used for the neutron diffraction measurements.⁶ The data reproduce earlier results on a different UCu₄Pd sample⁴ extremely well, exhibiting the rapid increase of linewidth with decreasing temperature and strong inhomogeneity in the low-temperature susceptibility (distribution width/mean susceptibility $\gtrsim 2$) predicted by disorder-driven theories of NFL behavior.^{2,8,9}

Positive muons (μ^+) from the M20 beam line at TRIUMF, Vancouver, Canada, were stopped in the sample and subsequently precessed in the sum of the applied field (if any) and the local internal field at the μ^+ site. As in previous studies,⁴ zero-field μ SR linewidths showed no evidence for spin freezing or static magnetism greater than $\sim 0.01\ \mu_B/\text{U}$ atom down to $\sim 2\text{ K}$.

The muon relaxation rate $1/T_1$ in low ($\sim 100\text{ Oe}$) fields applied parallel to the muon spin, which is due to U-moment thermal fluctuations,¹² was found to be unobservably slow ($\lesssim 10^4\text{ s}^{-1}$) at all temperatures. For the present case of coupling between U and μ^+ magnetic moments via a fluctuating dipolar field B_{dip} (Ref. 4), the U-moment fluctuation rate ν is given by¹² $1/T_1 = 2(\gamma_\mu B_{\text{dip}})^2/\nu$, where γ_μ is the μ^+ gyromagnetic ratio.

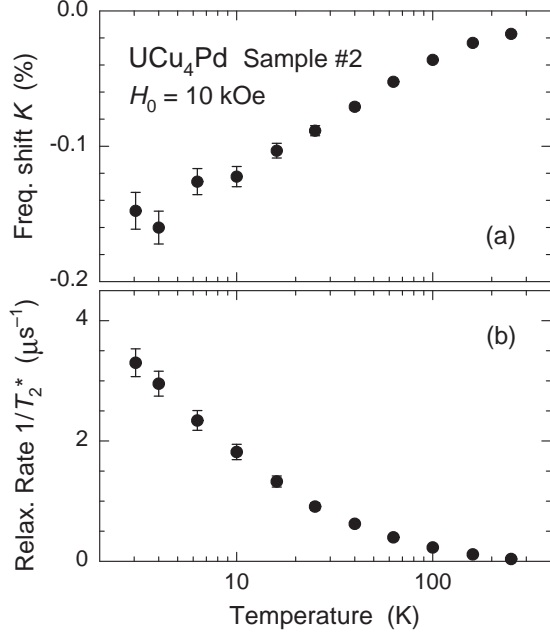


FIG. 3. Temperature dependence of μ^+ (a) frequency shift K and (b) relaxation rate $1/T_2^*$ in UCu_4Pd .

We find a lower limit on ν of $\sim 2 \times 10^{12} \text{ s}^{-1}$.

μSR spectra were obtained for applied fields H_0 between 2 and 20 kOe transverse to the μ^+ spin direction (TF- μSR) over the temperature range 3–300 K. A typical TF- μSR relaxation function is shown in Fig. 2, with the oscillatory factor removed by transforming to a reference frame rotating at the μ^+ precession frequency. It can be seen that the relaxation is exponential to within uncertainties. In spite of this the relaxation is not due to lifetime broadening, as evidenced by the negligible dynamic μ^+ relaxation rate discussed above. Instead, the exponential relaxation indicates a Lorentzian distribution of μ^+ Larmor frequencies.

Frequency shifts K and exponential relaxation rates $1/T_2^*$ were obtained from fits to the relaxation function

$$G(t) = Ae^{-t/T_2^*} \cos[\gamma_\mu H_0(1 + K)t + \phi] \quad (1)$$

to μ^+ time-differential relaxation data. Here A is the initial muon decay asymmetry, H_0 is the applied transverse field, and ϕ is the phase of the initial μ^+ spin orientation. The temperature dependence of K (not corrected for Lorentz and demagnetizing fields) and $1/T_2^*$ at $H_0 = 10 \text{ kOe}$ are shown in Fig. 3. Both these quantities increase considerably in magnitude with decreasing temperature. In the case of K this is expected, as both microscopic and macroscopic (Lorentz and demagnetization) contributions to K should be proportional to the bulk susceptibility.⁴ At 3 K no significant field dependence of K or of the half-width $\delta K = 1/(\gamma_\mu H_0 T_2^*)$ of the

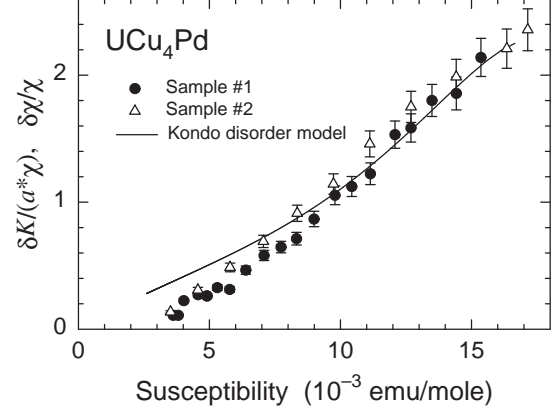


FIG. 4. Dependence of $\delta K/(a^*\chi)$ on bulk susceptibility χ , with temperature an implicit parameter, in UCu_4Pd . Symbols are defined in the text. Sample #1: previously-studied sample (Ref. 4). Sample #2: present sample, used in neutron diffraction studies (Ref. 6). Curve: $\delta\chi/\chi$ from the Kondo disorder theory (Refs. 3 and 4).

frequency shift distribution was observed for $H_0 < 20 \text{ kOe}$.

Estimates of the relative spread $\delta\chi/\chi$ were obtained using the relation

$$\frac{\delta\chi}{\chi} = \frac{\delta K}{a^*\chi}, \quad (2)$$

where a^* is an effective hyperfine coupling constant between U moments and μ spins.^{3,4} Assuming randomly-disordered inhomogeneity of the U-moment static susceptibility, a^* depends on the correlation length ξ_χ which describes the spatial variation of the inhomogeneity.³ It was found previously from comparison of TF- μSR and NMR linewidths^{3,4} that ξ_χ is of the order of a lattice parameter or smaller in $\text{UCu}_{5-x}\text{Pd}_x$, $x = 1.0$ and 1.5 . This can be seen qualitatively as follows:³ if ξ_χ were long (\gg a lattice parameter) the susceptibility in the vicinity of a given μ^+ site would be nearly uniform. Then the dipolar μ^+ hyperfine field would nearly vanish by symmetry at the $4a$ and $4d$ μ^+ sites found¹¹ in the end compound UCu_5 (cf. Fig. 1), which are octahedrally and tetrahedrally coordinated by U ions. For long ξ_χ , therefore, the distributed μ^+ shifts and hence the linewidth would be small, contrary to observation. The vanishing dipolar field also rules out any contribution to the powder-pattern linewidth due to anisotropy of the shift.

Figure 4 plots $\delta K/(a^*\chi)$ vs. the uniform bulk susceptibility, with temperature an implicit parameter. The data points were obtained from the μSR data and Eq. (2) using the dipolar-coupling value $\bar{a}^* = 0.096 \text{ molemu}^{-1}$ averaged over the two muon sites¹³ and calculated assuming short-range correlation.³ Data are shown for the

previously-studied⁴ sample (Sample #1) (reanalyzed for this comparison) and the present sample (Sample #2). It can be seen that the agreement between the two data sets is very good.

Also shown in Fig. 4 for comparison is $\delta\chi/\chi$ from the Kondo disorder model.³ The parameters defining the distribution function $P(T_K)$ were obtained from fits to the field and temperature dependence of the bulk susceptibility, and $\delta\chi/\chi$ was then calculated with no further adjustable parameters. Comparison with the disorder-driven Griffiths-phase theory (not shown), made in a similar way, is also satisfactory (cf. Ref. 9). Independently of agreement with a specific model, it is important to note that if the U-ion susceptibility were uniform (as in any NFL theory without disorder) $\delta K/(a^*\chi)$ would be constant;³ its strong dependence on χ and large low-temperature (large- χ) value (Fig. 4) are unambiguous evidence for susceptibility inhomogeneity.

It is conceivable that disorder plays no role in the susceptibility inhomogeneity, which then must have an ordered (periodic) spatial distribution. Such a periodically modulated susceptibility cannot be due to long-range spin ordering, since as discussed above zero-field μ SR shows that there is no static magnetism. We know of nothing to rule out a modulated susceptibility, but to our knowledge such behavior has never been observed without a corresponding modulated static moment (antiferromagnetism or a spin density wave).

Thus residual disorder appears to be the most likely explanation of these results. The reproducibility of $\delta K/(a^*\chi)$ between Sample #1 and Sample #2 (Fig. 4) is striking, however. The concentration of lattice defects in an arc-melted sample depends on conditions such as vapor pressures, cooling rate, etc., which are not likely to be reproduced exactly in two different laboratories. We speculate that the two samples may not have the same lattice defect concentration, in which case the inhomogeneity is remarkably insensitive to the amount of disorder. We also note that the low-temperature values of $\delta K/(a^*\chi)$ in UCu_4Pd and $\text{UCu}_{3.5}\text{Pd}_{1.5}$ (which is definitely a disordered alloy) are also very similar.⁴ These results raise the question of whether theories of NFL behavior which are *not* based on disorder can ever be verified in real compounds, which will always possess some level of defects. X-ray-absorption-fine-structure (EXAFS) and neutron pair-distribution-function (PDF) studies are under way to determine the amount of structural disorder in UCu_4Pd .

These results also provide important constraints on “cluster-based” mechanisms for NFL behavior (e.g., Ref. 9). First, the short correlation length discussed above seems to limit the number of f ions in the clusters to the order of a few. Second, the absence of observable μ^+ dynamic relaxation sets a lower limit of $\sim 2 \times 10^{12} \text{ s}^{-1}$ on the fluctuation rate of spins in clusters. Any cluster-based mechanism must therefore be consistent with the small size and rapid fluctuation rate of the clusters.

In the absence of evidence for a periodically modulated

susceptibility, we conclude that disorder-driven mechanisms are able to explain NFL behavior in UCu_4Pd . Further experimental work is needed to decide between various such mechanisms, and future NMR and μ SR experiments will be necessary to characterize disorder-driven NFL behavior in other nominally ordered compounds.

We are grateful to S. R. Dunsiger, M. Good, B. Hitti, R. F. Kiefl, and S. R. Kreitzman for assistance during the experiments, and to M. C. Aronson, W. P. Beyermann, C. H. Booth, A. H. Castro Neto, V. Dobrosavljević, J. M. Lawrence, A. J. Millis, R. Osborn, and J. D. Thompson for illuminating discussions. This research was supported in part by the U.S. National Science Foundation, Grants DMR-9418991 (U.C. Riverside), DMR-9510454 (Columbia), DMR-9705454 (U.C. San Diego), and DMR-9400755 (U. Florida), and by the U.C. Riverside Academic Senate Committee on Research, the Japanese agency NEDO (Columbia), and the Netherlands agencies FOM and NWO (Leiden), and was carried out in part under the auspices of the U.S. DOE (Los Alamos).

-
- * Present address: Lawrence Livermore National Laboratory, Livermore, California 94550.
 - ¹ See, for example, articles in *Proceedings of the Conference on Non-Fermi Liquid Behavior in Metals*, Santa Barbara, California, 1996, edited by P. Coleman, M. B. Maple, and A. J. Millis, J. Phys.: Condens. Matter **8** (1996).
 - ² O. O. Bernal, D. E. MacLaughlin, H. G. Lukefahr, and B. Andraka, Phys. Rev. Lett. **75**, 2023 (1995).
 - ³ D. E. MacLaughlin, O. O. Bernal, and H. G. Lukefahr, J. Phys.: Condens. Matter **8**, 9855 (1996).
 - ⁴ O. O. Bernal *et al.*, Phys. Rev. B **54**, 13000 (1996); D. E. MacLaughlin *et al.*, Physica (Amsterdam) **B230–232**, 606 (1997).
 - ⁵ B. Andraka and G. R. Stewart, Phys. Rev. B **47**, 3208 (1993).
 - ⁶ R. Chau, M. B. Maple, and R. A. Robinson, Phys. Rev. B **58**, 139 (1998).
 - ⁷ K. Matsuhira, T. Sakakibara, and H. Amitsuka, Physica B **206 & 207**, 326 (1995).
 - ⁸ E. Miranda, V. Dobrosavljević, and G. Kotliar, J. Phys.: Condens. Matter **8**, 9871 (1996); Phys. Rev. Lett. **78**, 290 (1997).
 - ⁹ A. H. Castro Neto, G. Castilla, and B. A. Jones, cond-mat/9710123 (unpublished).
 - ¹⁰ M. C. de Andrade *et al.*, cond-mat/980281 (unpublished).
 - ¹¹ S. R. Barth *et al.*, Hyperfine Interact. **31**, 397 (1986); S. R. Barth *et al.*, J. Magn. Magn. Mater. **76 & 77**, 455 (1988); A. Schenck *et al.*, Hyperfine Interact. **64**, 511 (1990).
 - ¹² A. Schenck, *Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics* (A. Hilger, Bristol & Boston, 1985).
 - ¹³ the values of a^* for the two sites differ from this average by $\sim 10\%$, which does not affect the conclusions of this paper.